

**Claims:**

1. A method for producing aligned carbon nanotubes and/or  
5 nanofibres comprising providing finely divided substrate particles having substantially smooth faces with radii of curvature of more than 1  $\mu\text{m}$  and of length and breadth between 1  $\mu\text{m}$  and 5 mm and having catalyst material on their surface and a carbon-containing gas at a temperature and pressure at which the carbon-containing gas will react to form carbon when in the presence of the supported catalyst, and forming aligned nanotubes and/or nanofibres by the carbon-forming reaction.  
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- 15 2. A method as claimed in Claim 1, wherein the faces have radii of curvature of more than 10  $\mu\text{m}$ .
3. A method as claimed in either one of the preceding  
20 claims, wherein the substantially smooth faces are substantially flat.
4. A method as claimed in any one of the preceding claims wherein the catalyst material is dispersed in clusters  
25 on the surface of the substrate particles.
- 30 5. A method as claimed in Claim 4, wherein the catalyst material clusters are from 0.5 nm to 100 nm in dimension.

6. A method as claimed in Claim 5, wherein the catalyst material clusters are from 3 nm to 50 nm in dimension.
7. A method as claimed in any preceding claim, wherein the substrate particles having catalyst material on their surface are prepared by depositing catalyst material on the surface of the substrate particles.  
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8. A method as claimed in Claim 7, wherein the catalyst material is deposited by electroless deposition, solvent drying, supercritical drying, sputtering, physical vapour deposition or electroplating.  
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9. A method as claimed in any one of Claims 1 to 6, wherein the substrate particles having catalyst material on their surface are prepared by providing finely divided substrate particles and a catalyst precursor material and decomposing the catalyst precursor material to form the catalyst material in the presence of the substrate particles such that the catalyst material is deposited on the substrate particles.  
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10. A method as claimed in Claim 9, wherein the catalyst precursor material is decomposed while the substrate particles are in contact with the carbon-containing gas.  
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11. A method as claimed in any one of Claims 1 to 6, wherein the substrate particles having catalyst material on their surface are prepared by providing finely divided substrate particles of a material that is decomposable  
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to form catalyst material on the surface of non-catalyst substrate particles and decomposing said material.

12. A method as claimed in any preceding claim, wherein the  
5 length and breadth of the substrate particle faces are  
between 10  $\mu\text{m}$  and 500  $\mu\text{m}$ .

13. A method as claimed in any preceding claim, wherein the  
substrate particles are of silica, alumina, carbon,  
10 mica, magnesium oxide, calcium oxide, sodium chloride,  
pure metal or aluminosilicate or a mixture of two or  
more thereof.

14. A method as claimed in Claim 13, wherein the substrate  
15 particles are of graphite, aluminium or titanium.

15. A method as claimed in any preceding claim, wherein the  
substrate particles are anisotropic.

20 16. A method as claimed in Claim 15, wherein the substrate  
particles have one dimension larger than the other two  
dimensions or wherein the substrate particles have one  
dimension smaller than the other two dimensions.

25 17. A method as claimed in any one of preceding claims where  
the substrates are coated with a buffer layer.

18. A method as claimed in any preceding claim, wherein the  
substrate is freshly prepared.

19. A method as claimed in Claim 18, wherein the substrate is prepared by colloidal processing, spray-drying, hydrothermal processing, or ball-milling.

5 20. A method as claimed in any preceding claim, wherein the catalyst material is a transition metal, an alloy of two or more thereof, a compound of a transition metal or a mixture of two or more compounds of transition metals.

10 21. A method as claimed in Claim 20, wherein the transition metal is iron, cobalt, molybdenum or nickel.

15 22. A method as claimed in Claim 9 or Claim 10, wherein the catalyst precursor is a transition metal carbonyl, or a transition metal cyclopentadienyl compound.

20 23. A method as claimed in Claim 22, wherein the catalyst precursor is ferrocene, nickelocene, cobaltocene, iron pentacarbonyl or nickel tetracarbonyl.

25 24. A method as claimed in any preceding claim, wherein the carbon containing gas is carbon monoxide, an oxygen containing organic compound or a hydrocarbon, or a mixture of two or more thereof.

25 25. A method as claimed in Claim 24, wherein the carbon containing gas is carbon monoxide, benzene, toluene, xylene, cumene, ethylbenzene, naphthalene, phenanthrene, anthracene, methane, ethane, propane, hexane, ethylene, propylene, acetylene, formaldehyde, acetaldehyde, acetone, methanol, ethanol or a mixture of two or more thereof.

26. A method as claimed in any one of the preceding claims wherein one or more boron and/or nitrogen containing compound is provided in addition to the carbon containing gas.
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27. A method as claimed in any one of the preceding claims, wherein one or more promoter compounds is provided in addition to the carbon containing gas.
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28. A method as claimed in Claim 27, wherein the promoter compound is thiophene.
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29. A method as claimed in any one of the preceding claims, wherein a diluent gas is provided mixed with the carbon containing gas.
30. A method as claimed in any preceding claim, wherein the substrate particles are reacted within a fluidised bed.
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31. A method as claimed in any preceding claim, wherein substrate particles are provided and product particles are removed from a reaction vessel in a continuous fashion.
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32. A method as claimed in any preceding claim, further comprising the step of recovering the aligned nanotubes and/or nanofibres.
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33. A method as claimed in any one of the preceding claims, wherein gaseous effluent from the reaction is recycled with or without clean up.

34. A method as claimed in any preceding claim, wherein the reaction takes place at a temperature between 650 °C and 1250 °C.

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35. Finely divided substrate particles having substantially smooth faces with radii of curvature of more than 1 µm and of length and breadth between 1 µm and 5 mm with a catalyst material on the surface of the substrate particles and with aligned carbon nanotubes and/or nanofibres on the surface of the substrate particles.

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36. Carbon nanotubes and/or nanofibres produced by a method as claimed in any one of the preceding claims.

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37. Nanotubes and/or nanofibres as claimed in Claim 36, wherein the nanotubes and/or nanofibres are separated from the substrate particles by partial or complete dissolution of the substrate particles or catalyst materials.

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